

# The effect of CO<sub>2</sub> injection on caprock permeability in deep saline aquifers

Dinesha Wanigarathna Jayasekara

*Department of Civil Engineering, Monash University, Clayton, Australia*

[dinesha.wanigarathnajayasekara@monash.edu](mailto:dinesha.wanigarathnajayasekara@monash.edu)

Ranjith Pathegama Gamage<sup>1</sup>

*Department of Civil Engineering, Monash University, Clayton, Australia*

[ranjith.pg@monash.edu](mailto:ranjith.pg@monash.edu)<sup>1</sup>

**ABSTRACT:** Investigating the caprock permeability in deep saline aquifers is crucial to identify the risks of CO<sub>2</sub> back-migration. The caprock is subjected to geochemical reactions which can alter the leakage pathways. Thus, it is crucial to identify the supercritical CO<sub>2</sub> (scCO<sub>2</sub>) flow behaviour via fractures in caprock and its permeability to estimate the permanence of injected CO<sub>2</sub>. Therefore, the objective of the current study is to find the effect of scCO<sub>2</sub> flow on fractured caprock permeability. For that, a fractured siltstone sample was saturated in deionized water and conducted scCO<sub>2</sub> permeability tests using a high-precision advanced core flooding apparatus under different injection pressures and confinements. After that, the siltstone sample was saturated in 10 % w/w NaCl brine and conducted scCO<sub>2</sub> permeability tests as described earlier. The results show that the brine-saturated sample has low permeability compared to water-saturated siltstone sample. The reason would be the deposition of evaporites during scCO<sub>2</sub> flow through the fractured sample. This is known as CO<sub>2</sub> dry-out phenomenon or absorbing moisture into the scCO<sub>2</sub>, making the remaining brine saturated with salts. Thus, the CO<sub>2</sub> back-migration through the caprock discontinuities becomes minimized due to CO<sub>2</sub> dry-out phenomenon, which is an advantage for the caprock integrity in deep saline aquifers. In addition, the aquifers with high salinity contents show significant dry-out phenomenon because pore fluid easily gets supersaturated with salts due to evaporation of moisture into the scCO<sub>2</sub>.

**Keywords:** CO<sub>2</sub> sequestration, caprock, fracture, scCO<sub>2</sub>, permeability

## 1. Introduction

Since the industrial revolution, the atmospheric concentration of CO<sub>2</sub> and other greenhouse gases has been risen up, leading to global warming and climatic changes, affecting weather patterns, flora and fauna [1-5]. The new Victorian state temperature record (in Mildura) in September 2017 with a top of 37.7°C, provides the most recent evidence to prove the adverse effects caused by the atmospheric CO<sub>2</sub>. Accordingly, the modern world is experiencing calamitous side effects such as rising of sea levels, polar ice melting, changes in global water and carbon cycles [4]. For instance, the Great Barrier Reef: the world's largest reef system has currently been subjected to a significant threat by being a victim of climatic change caused ocean acidification and the rise of sea temperature. The Great Barrier Reef Marine Park Authority (GBRMPA), Australia, reveals that the sea temperature rise in the year 2016 caused the worst destruction of coral reefs, with the highest recorded coral bleaching in the documented history of the Great Barrier Reef. In addition, the dry condition and high temperatures due to climate change can create devastating catastrophes such as British Columbia's wildfire, which destroyed 1,079,569 acres of land on July 6, 2017. Therefore, it is evident that the rise of anthropogenic CO<sub>2</sub> levels in the atmosphere is not only a threat to the human lifestyle but also for the survival of flora and fauna. The atmospheric CO<sub>2</sub> concentration will exceed 50 billion tonnes per year by 2050 if it is not controlled by any means [6]. Therefore, it is essential to yield mitigatory measures to reduce climate change and

to achieve a sustainable living environment. In other words, it is essential to implement appropriate CO<sub>2</sub> emission control techniques to create a better place for living beings.

As a solution, carbon capture and storage (CCS) techniques have been gained special attention due to its high efficiency and long-term stability [7]. Among, sequestering CO<sub>2</sub> in deep saline aquifers is one of the most promising CCS techniques in the world due to its many advantages such as extensive availability, high efficiency, long-term stability, quick and simple capture process, huge storage capacity and advantageous locations close to major CO<sub>2</sub> sources [8]. The injected CO<sub>2</sub> in deep saline aquifers lie in supercritical phase due to the temperature and pressure of the aquifer, which are greater than the critical point of CO<sub>2</sub>. Caprock, one of the major components of deep saline aquifers plays important roles in CO<sub>2</sub> sequestration process such as preventing back-migration of CO<sub>2</sub>, limiting seismic events, inhibiting contamination of CO<sub>2</sub> with fresh underground water, restraining ground heave, and contributing for mineral trapping [9]. However, one of the major concerns of underground CO<sub>2</sub> sequestration in deep saline aquifers is the back-migration of CO<sub>2</sub> through the caprock (mainly depends on caprock permeability) due to its degradation as a result of the dissolution of minerals. Thus, it is really crucial to investigate the caprock permeability that highly depends on mineral reactivity due to carbonic acid created by injected CO<sub>2</sub>. Hence, the objective of this study is to investigate the caprock permeability due to CO<sub>2</sub> injection in deep saline aquifers.

## 2. Methodology

Siltstone samples obtained from Eidsvold basin in Queensland were used as representative of caprock. The composition of the rock is 35% quartz, 53% kaolinite, 8% muscovite, 3% alunite and 1% other minerals [10]. The cylindrical siltstone sample (38 mm diameter and 76 mm long) was cored, cut and ground in Deep Earth Energy Research Laboratory (DEERL) at Monash University, according to ASTM standards (D2664). The prepared siltstone sample was oven-dried at 40 °C for 5 days to remove all moisture trapped in the rock pores and such kind of low temperature was used to avoid appearing thermal cracks. After that, the sample was subjected to Brazilian test as shown in Fig. 1 to make a fracture along the cylindrical sample. The fractured sample was subjected to computed tomography (CT) scanning to identify the fracture volume using the Australian Synchrotron radiation facility. The digital image processing was conducted using Avizo 9.4.0 software and the extracted fracture is shown in Fig. 2. Firstly, the median filter and then unsharp mask filter were applied to enhance the quality of the CT images. After that, the image segmentation was conducted to separate rock matrix, pores and rock fracture.



Figure 1: Brazilian test conducted to make a fracture in the sample

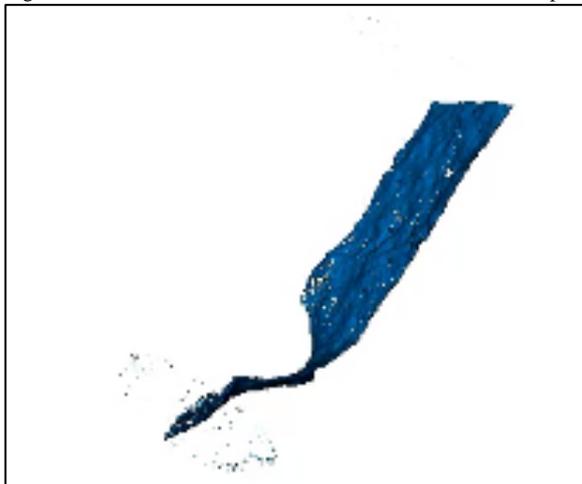


Figure 2: The extracted fracture using CT scanning

After that, the fractured sample was saturated in water until it reaches 100 % degree of saturation using a desiccator under vacuum conditions as shown in Fig.3. Usually, within one month the sample becomes saturated with fluid, but in order to make sure its fully saturation, the sample was saturated for 3 months. After that, the permeability tests were conducted using a high-precision advanced core flooding apparatus as shown in Fig. 4. It can simulate the complex underground conditions by applying different temperatures, confining and injection pressures. The temperature is applied using an electric oven which can be heated from 20 to 150 °C and the sample is confined using water. In addition, different kinds of fluids (liquids and gases) such as water, brine, CO<sub>2</sub> and N<sub>2</sub> can be injected into the samples.

In order to inject CO<sub>2</sub> in the supercritical state, the gas was injected at different pressures and temperature which are greater than the critical point of CO<sub>2</sub> such that injection pressures were 8, 9 and 10 MPa and the temperature was maintained at 35 °C. The permeability tests were conducted under four confining pressures such as 15, 20, 25 and 30 MPa. After that, the sample was saturated in 10 % w/w NaCl brine sample for 3 months in a desiccator and conducted scCO<sub>2</sub> permeability tests as described earlier. The permeability was calculated using a transient method called pore pressure transmission method as shown in Eq. (1) and (2) [11]. The speciality of this method is the consideration of porosity to calculate permeability since siltstone has high permeability compared to granite. Thus, the pore pressure transmission method was used rather than calculating using conventional pulse decay method. Here, infinite storage was created in the upstream by injecting scCO<sub>2</sub> at a constant flow rate while measuring the downstream pressure development by closing the downstream pressure release valve.



Figure 3: Desiccator used to saturate the siltstone sample



Figure 4: The core flooding apparatus in Monash University

$$K = \frac{\alpha \mu L^2 \beta_f \varphi}{-\theta^2} \quad (1)$$

$$\tan \theta = \frac{\gamma}{\theta} \quad (2)$$

Where,  $\beta_f$  is the fluid compressibility;  $K$  is the permeability;  $\mu$  is the dynamic viscosity,  $L$  is sample length  $\varphi$  is the porosity (fracture volume) and  $\gamma$  is the ratio of pore space volume of the sample to downstream volume.

### 3. Results and discussion

The calculated transient permeability values for water-saturated siltstone sample under different confining pressures are shown in Fig. 5. Accordingly, the permeability increases with increasing injection pressures because the effective stresses reduce at high pore pressures. Moreover, the permeability values reduce at high confinement due to closure of flow paths as a result of high stress created on the rock sample. Fig. 6 shows the scCO<sub>2</sub> permeability values for brine saturated sample under different confinements and injection pressures. Accordingly, the scCO<sub>2</sub> permeability of brine-saturated sample is lower than water-saturated permeability values. For instance, the CO<sub>2</sub> permeability for water-saturated sample under 15 MPa confinement at 8 MPa injection is 0.52  $\mu$ D while for brine-saturated sample is 0.03  $\mu$ D. Therefore, there is 94 % CO<sub>2</sub> permeability reduction. The reason is subflorescence phenomenon due to CO<sub>2</sub> dry-out effect or the deposition of evaporites in the flow paths as a result of absorption of moisture available in rock pores into flowing scCO<sub>2</sub> [12]. Thus, the permeability of the fractures in caprock reduces due to scCO<sub>2</sub> flow and eventually it can reduce the risk of back-migration of injected CO<sub>2</sub> which is favourable for maintaining the caprock integrity. Moreover, the caprock integrity can be maintained very well in deep saline aquifers if the pore fluid salinity is high since it can create a significant CO<sub>2</sub> dry-out phenomenon [10] which eventually reduce the flowability of injected CO<sub>2</sub> through caprock discontinuities.

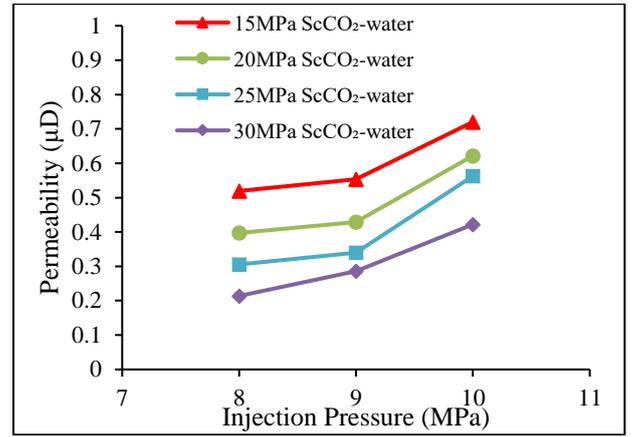


Figure 5: The scCO<sub>2</sub> permeability of the fractured siltstone sample which is saturated with water

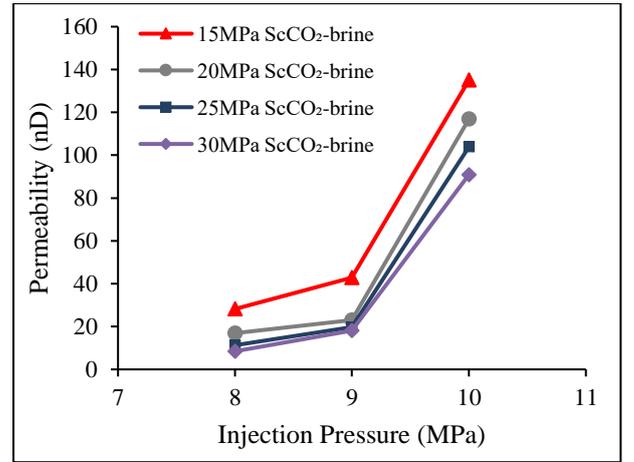


Figure 6: The scCO<sub>2</sub> permeability of the fractured siltstone sample saturated with brine

### 4. Conclusion

The caprock in deep saline aquifers can be subjected to chemico-mechanical changes due to the injected CO<sub>2</sub>. As a result, the caprocks with fractures can be appeared (due to mineral dissolution and pressure changes) which can induce CO<sub>2</sub> back-migration risks. Thus, the CO<sub>2</sub> permeability in fractured caprock was investigated in the current study using CT scanning and high-precision advanced core flooding apparatus. For that, CO<sub>2</sub> permeability tests for both water-saturated and brine-saturated samples were conducted to compare the flow ability.

According to the results, the scCO<sub>2</sub> permeability of the fractured caprocks in deep saline aquifers is low due to CO<sub>2</sub> dry-out phenomenon, which is an advantage for the caprock. In other words, CO<sub>2</sub> dry-out phenomenon is the evaporation of moisture into the scCO<sub>2</sub> making the remaining brine super-saturated with salts. Thus, salt crystallization occurs during scCO<sub>2</sub> flow through the fractures in caprock. Moreover, the dry-out phenomenon becomes very significant if the salinity of the pore fluid is high. Thus, the caprock integrity can be maintained very well in an aquifer which contains high salinity pore fluids.

However, there are different types of ions in pore fluid other than Na<sup>+</sup> and Cl<sup>-</sup> in brine. Therefore, it is crucial to find the influence of those ions on salt crystallization during scCO<sub>2</sub> flow. Therefore, such kind of permeability

study is recommended as a future work along with chemical analysis to predict the influence of ions on CO<sub>2</sub> dry-out phenomenon.

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